Effective coatings improve the performance of any device designed to emit or collect light over a range of angles and wavelengths. Improved broadband and wide angle antireflection coatings (ARCs) are a simple and direct way to improve solar cell performance. In this paper we demonstrate a multilayered ARC optimized using a new meta-heuristic algorithm called the average uniform algorithm (AUA). Comparison between the well-known genetic algorithm and the AUA showed that both achieved similar results but the AUA converged much faster. The coating optimized by AUA for broadband and wide-angle emission is applied to a copper zinc tin sulfide based thin film solar cell by co-sputtering of high and low refractive index material. A significant improvement in efficiency was observed over wide angle and bandwidth with a typical improvement of 15% over the uncoated solar cell. This AUA methodology is proven to be an efficient method for design of general ARCs. © 2014 Optical Society of America

1. Introduction
Antireflection coatings (ARCs) are a crucial part of solar cells, or of any technology that is designed to collect or emit light with highest efficiency. Good coatings can significantly increase the solar power harvested. Much research has been done on effective ARCs for solar cells and many different topologies of ARCs have been studied in order to find one with the optimal properties.

Design of optical coatings is a complicated problem. It is easy to calculate the transfer function of a dielectric, but hard, in general, to design a dielectric stack with a particular reflectivity or transmissivity. Many different heuristic methods have been applied. In this paper, we discuss a new and effective method for coating design, and demonstrate a coating design with it as a proof of concept.

The simplest ARC is a quarter-wave thick layer of a transparent material having a refractive index \( n_1 \) as the geometric mean of that of the material on either side of the coating, \( \sqrt{n_0n_2} \), where \( n_0 \) and \( n_2 \) are the refractive indices of incident and substrate material. In principle this achieves zero reflectivity, but at only one specific angle and wavelength, and requires a material with the correct intermediate refractive index \( n_1 \). Many applications, particularly solar cells, have to handle light over a wide range of wavelengths and incident angles.

A widely used technique other than quarter-wave thick monolayer or double layer coating is gradient index (GRIN) coatings. These coatings are designed to avoid the interference effect due to the reflection
from multiple interfaces with different refractive indices on either side. GRIN coatings achieve low reflectivity over a wide spectral and angular range through gradual index changes. After GRIN ARC films were experimentally realized by Jacobsson and Martensson in the 1960s [1], Southwell showed results for different index profile of GRIN coating, concluding that a quintic-type distribution of index profile is best [2]. Ge et al. showed more than 10% improvement for normal incidence in power conversion efficiency of Cu$_2$SnZnS$_4$ (CZTS)-based thin film solar cell with linear GRIN coating from cosputtered aluminum-doped zinc oxide (AZO) ($n = 1.9$) to SiO$_2$ ($n = 1.5$) [3].

Schubert et al. compared the performance of GRIN coatings with ARC using multiple discrete layers. It was shown that, in practical cases where refractive index choices are constrained, discrete ARCs can surpass the performance of continuously graded coatings by taking advantage of interference effects. It was also shown that the coating offered substantially lower reflectivity than even a hypothetical, infinitely thick continuously graded coating [4].

Because typically materials are not available for optimal GRIN coatings for most practical applications, many researchers are working on optimizing multiple discrete layer coatings. Genetic algorithms (GAs) have been explored for different numbers of layers by many authors [5,6]. Chang and Chen used a simulated annealing algorithm for optimization of a multilayer coating for silicon and CuIn$_x$Ga$_{1-x}$Se$_2$ (CIGS)-based cells and showed improvement over GAs [5]. Other authors have tried surface texturing methods including moth eye, pyramids, and other types of nanostructuring [7–9]. Jheng et al. reported that by controlling the morphology of ZnO nanostructures on top of a CZTS solar cell, the energy conversion efficiency increased from 4.8% to 5.3% [10].

The approach that we discuss in this paper is a new meta-heuristic approach, called the average uniform algorithm (AUA), to obtain an optimum ARC profile with given constraints. The result is compared with the most common meta-heuristic approaches, the GAs [11–13].

With the basic GA method, an initial population of random arrangements of layer indices and thicknesses is generated, and a “fitness function” (defined as the reflectivity over a set of wavelengths and angles) is calculated for all members of the population. Those with the best fitness functions are kept and modified to evolve to the next generation of coatings. The refinement of the AUA algorithm is that it uses the average of only the best members to define the new sample set. It also generates some new random coatings each generation to avoid getting locked into a local minimum. This approach has been previously proposed and applied to classic optimization problems, such as Himmelblau’s function and the six-hump camel back function [14]; here, we apply it to a very practical problem of realizing an effective ARC. This small variation enables it to converge at least 10 times faster than the conventional GA to a better solution.

Once designed, the coatings are fabricated and applied to the CZTS solar cell to validate its efficacy. Power efficiency improvements in the range of 12%–36% were achieved at different angles of incidence, and are presented in the results and discussion section.

2. Methodology: Optimization

The process for designing an optimized coating, using both the AUA and conventional GA, is outlined below.

Both of them begin with a population of 100 random solutions generated from a continuous random distribution. Each individual solution consists of 10 different refractive indices ($n_n$) with their respective thicknesses ($t_n$). The objective function here is to minimize the reflectivity $R$ of the 10-layer stack:

$$ R = f(x_1, x_2, x_3, x_4, x_5, x_6, x_7, x_8, x_9, x_{10}), $$

where

$$ x_n = (n_n, t_n), $$

and

$$ n_n \in [n_{\min}, n_{\max}] \quad \text{and} \quad t_n \in [t_{\min}, t_{\max}]. $$

The fitness function or figure of merit (FOM) for each member of the population is defined to be the sum of the reflectivity over the selected set of angles (from 0° to 60°) and wavelengths (from 350 to 950 nm); for this definition, a smaller number is better. The reflectivity is calculated for each member of the population using a transfer matrix method.

The population is then sorted in ascending order of the FOM. The optimization algorithm then obtains the population for the next generation based on the current generation, and this process continues iteratively, generation by generation, until the terminating conditions are met. In the classic GA, the next generation is determined from only the fittest candidates of the current generation and their mutations. The AUA method includes some new, randomly generated members, as well, whose characteristics are centered around the average of the fittest members of the population. These will be discussed in detail in the following subsections.

For this proof-of-concept realization of an AUA-designed coating, the allowable indices in the optimization are kept between 1.93 and 1.50. The maximum allowable thickness/layer is held to <40 nm (total thickness of <400 nm) to constrain it to a realistic coating and minimize absorption losses.

A. Genetic Algorithm

Since a GA imitates the biological evolution process to create the new generation, it uses selection, crossover, and mutation to obtain the next generation:
- 10% of the best sample are carried over to the next generation;
- 40% are created by crossover or recombination; and
- 50% are created by mutation.

### B. Average Uniform Algorithm

AUA is a mathematical approach that tries to progressively narrow the search space to achieve the optimum result. This algorithm was proposed to solve a nonconvex and multimodal continuous optimization problem, and is thus useful for problems with more than one local solution. The algorithm is described in more detail elsewhere [14]. In sum:

- 80% of new population is created by creating a progressively narrower search space window centered around the average of a few of the best solutions from the last generation. This helps in exploiting the search space neighboring the best solutions of the last generation; and
- 20% of the new population is created from the uniform random distributed numbers in the original sample space. This allows for further exploration of search space for global minima.

The modification of adding some random members each generation allows the algorithm to find other solutions that may have not happen to be randomly generated in the initial random population. The only operations done on the current population are evaluating, sorting, and averaging. While this may not be quite as flexible as a GA in other problems, for this specific problem, the dependency of reflectivity on properties is such that averaging of the best solutions does converge much faster than a conventional GA.

### 3. Methodology: Fabrication

ARCs have a significant role in the photovoltaic industry. Many coatings are designed for crystalline-silicon-based cells [15], which have smooth surfaces. However, a number of solar devices collect the light through a transparent conductive oxide [such as AZO or indium tin oxide (ITO)] on the top surface. These cells are polycrystalline and rougher than silicon-based cells.

Here, as a proof of concept, we realize a solar cell coating optimized for a CZTS solar cell with an AZO top contact layer, whose detailed fabrication is described elsewhere [16]. As the quantum efficiency of the cell is not the same throughout the band, the optimizing function is weighted by the quantum efficiency of the CZTS solar cell at that wavelength. This particular cell, while not state-of-the-art for CZTS, is sufficient to evaluate the effect of the AR coating on performance.

The materials used to sputter the coating are co-sputtered AZO (with $n = 1.93$) and silicon dioxide (SiO$_2$) ($n = 1.5$). For each slice determined by the AUA, the specific ratio of sputtering rates for AZO and SiO$_2$ are computed to realize that index. That layer is then sputtered for the calculated time to achieve that thickness. Other authors have shown that the refractive index of similar mixed oxide materials, either cosputtered [17] or deposited in very thin layers [18], is, within a few percent, linearly proportional to the volume fraction of the deposited materials [19]. Hence, the assumed refractive index is taken as proportional to the composition fraction of each material for modeling and analysis.

Cosputtering was done at a base pressure of less than $10^{-5}$ Torr. The substrate was not heated during or after the deposition.

### 4. Results and Discussion

The AUA and GA were given the same constraints of materials and thicknesses and were used to optimize the reflectivity over a range of angles from 0° to 60°, and wavelengths from 350 to 950 nm, constrained with a thickness per layer <40 nm (overall thickness <400 nm), and indices between 1.50 and 1.93.

The optimization conditions capture the requirements on solar cells, which harvest energy over a broad wavelength range over many angles throughout the day. The constraints on thickness and materials were to realize a practical coating, less than 400 nm thick, that could be fabricated from sputtered AZO and SiO$_2$.

As can be seen from Fig. 1, the AUA converges very fast compared to the GA. The AUA takes less than 100 generations to reduce the fitness function to under 26, while the GA takes more than 1000 generations.

The specific coatings calculated with the standard GA and the AUA using the same constraints on allowed thickness and refractive index are shown in Fig. 2. Other authors have optimized systems constrained to be two layers, or three layers [6]; here, we are providing 10 independent layers for optimization, which the algorithm can make distinct or similar.

To understand the effect of the constraint on film thickness on ARC, we simulated the algorithm with the constraint of maximum thickness ($t_{\text{max}}$) ranging from 40 to 2000 nm. Ten different runs are presented in Table 1. In all these runs, AUA comes up with better fit value, proving its consistency. The calculated reflectivity for several different films is shown in

![Figure 1](image-url)  
**Fig. 1.** Convergence of GA and AUA.
Fig. 2. Index profile of optimized coating from AUA and GA.

**Table 1. FOM of GA and AUA for Different Runs**

<table>
<thead>
<tr>
<th>Run</th>
<th>Parameter Changed</th>
<th>FOM of GA</th>
<th>FOM of AUA</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( t_{\text{max}} = 40 )</td>
<td>13.59</td>
<td>13.33</td>
</tr>
<tr>
<td>2</td>
<td>( t_{\text{max}} = 50 )</td>
<td>13.24</td>
<td>13.25</td>
</tr>
<tr>
<td>3</td>
<td>( t_{\text{max}} = 60 )</td>
<td>13.37</td>
<td>13.23</td>
</tr>
<tr>
<td>4</td>
<td>( t_{\text{max}} = 70 )</td>
<td>13.59</td>
<td>12.56</td>
</tr>
<tr>
<td>5</td>
<td>( t_{\text{max}} = 80 )</td>
<td>13.35</td>
<td>11.97</td>
</tr>
<tr>
<td>6</td>
<td>( t_{\text{max}} = 90 )</td>
<td>13.05</td>
<td>12.8</td>
</tr>
<tr>
<td>7</td>
<td>( t_{\text{max}} = 100 )</td>
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<td>12.13</td>
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<td>8</td>
<td>( t_{\text{max}} = 110 )</td>
<td>12.78</td>
<td>12.14</td>
</tr>
<tr>
<td>9</td>
<td>( t_{\text{max}} = 120 )</td>
<td>12.41</td>
<td>12.32</td>
</tr>
<tr>
<td>10</td>
<td>( t_{\text{max}} = 130 )</td>
<td>12.64</td>
<td>12.37</td>
</tr>
</tbody>
</table>

Fig. 3. The overall thickness and reflectivity are similar for different ranges of thickness constraint. For example, for all total thicknesses constrained to be less than 400 nm, very similar optimized coatings were determined, with an optimum thickness of around 170 nm. If the algorithm were allowed to go over 400 nm, it achieved a qualitatively different minimum with an optimum thickness of around 440 nm, as shown in Fig. 3. Also shown in Fig. 4 for comparison is the reflectivity of a GA optimized and AUA optimized ARC at normal and 60° angle of incidence.

Although the reflectivity did decrease with increasing thickness, film absorption increases as well. As a practical compromise the film thickness fabricated is kept under 200 nm. Comparing the optimized results (curves B and C) with the monolayer coatings (curve A) shows clearly the reduced reflectivity this multilayer coating over a broad spectrum and a wide angle range.

The designed coating was first applied to AZO-coated glass and the result is shown in Fig. 5. The resulting specular reflectance spectrum closely matches the simulated spectrum, suggesting that the designed layer was successfully realized. There are some discrepancies in simulated transmission because the simulation does not model the absorption from the layers. A maximum of 18% improvement in transmission was seen at 443 nm, and specular reflection was measured as low as 0.01% at several wavelengths. The average improvement of 62.7% in reflectance and 6.9% in transmission was seen between 350 and 950 nm.

The same coating was applied to the CZTS solar cell. Because the surface of the solar cell is rough...
due to the polycrystalline nature of the absorber, the diffusive reflection is quite significant, and much greater than the specular reflectance measured from the glass. Figure 6 shows the diffuse reflectance of the solar cell at normal incidence, measured in an integrating sphere. The average diffuse reflectance is reduced from ∼9% to ∼2% over the band from 350 to 950 nm after the application of the coating.

To verify the ARCs improvement in transmittance to the CZTS solar cell, the external quantum efficiency (EQE) of the solar cell before and after coating was measured with a small focused light spot, using a quantum efficiency measurement system from PV Measurements Inc. equipped with a xenon light source and a monochromator with a chopper. As shown in Fig. 7, a maximum increase of 9.8% (82.3%–90.4%) at around 590 nm was achieved.

The improvement in EQE and in reflection were compared as follows: the internal quantum efficiency (IQE) of the solar cell was approximated by the EQE of the cell after ARC. With that assumption, the EQE average increase in percentage points (3.4%) was roughly equal to the improvement in reflection, in percentage points, times the IQE of the solar cell, which was improved an average of 3.5%. The curves of measured EQE and calculated expected improvement matched well across the spectrum.

Figure 8 shows the power conversion efficiencies measured at different angles. The measurement was done using a Photo Emission Tech solar simulator under air mass (AM) 1.5 for terrestrial application conditions from 0° to 70° with a 10° interval. The improvement in short circuit current ($I_{sc}$) corresponding to the improved light harvesting caused by the ARC is the main factor in the solar cell efficiency improvement. The efficiency improvement was 14.3% (3.63%–4.15%) at normal incidence and up to 35.8% (1.06%–1.44%) at the highest angle measured (70°). The percentage improvement is significantly better than prior results achieved through a coating constrained to a linear graded topology, which realized a power efficiency improvement of 10.7% at normal incidence and 24.4% at 70° [3].

5. Conclusion

A multilayer ARC was optimized for a CZTS-based solar cell using GA- and AUA-based meta-heuristic algorithms. Both the algorithms achieved similar optimum coatings, but the AUA converges to the best result about 10 times faster than with GA. For practical algorithmic designs, where it is desirable to quickly optimize complex coating topologies, this could be a powerful new tool.

The multilayer coating applied to the CZTS solar cell improved the EQE up to 9.8%. The power conversion efficiency showed significant improvements over a wide range of angles, with maximum of 35.8% at the highest angle measured at 70°. At normal incidence, which is of most concern, it realized a 14.3% improvement.

The modest increase in solar cell efficiency is limited by the intrinsic efficiency of the material; much greater absolute efficiency increases would be expected for those solar cells with higher intrinsic efficiency. The same algorithm can be applied for various applications with different reflection and transmission requirements, such as optical filters.
The results here are partly limited by the choice of SiO$_2$ as the low refractive index material. With the availability of new, lower refractive index material, it is anticipated that the results reported here can be further improved significantly.

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References